Spark Plasma Sintering of Diamond-Reinforced Uranium Dioxide

Composite Fuel Pellets

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Abstract

In an effort to increase the thermal conductivity of UO₂ fuel pellets, high density UO₂-5 vol% diamond composite pellets were fabricated using the spark plasma sintering (SPS) technique. Diamond particles with nano-size (0.25 μm) and several micro-sizes (3 μm , 12 μm and 25 μm) were mixed with UO₂ powder and sintered using SPS at 1300°C-1600°C with a hold time of 5 minutes. The resultant density, chemical reaction, microstructure, thermal conductivity and Young's modulus of the sintered pellets were investigated. The pellets with 3 μm diamond particles had a uniform distribution of particles as well as better thermal and mechanical properties compared to others. An increase in thermal conductivity of up to 41.6%, 38.3% and 34.2% at 100°C, 500°C and 900°C, respectively, was measured in the UO₂-diamond composite pellets compared to the pure UO₂ fuel pellets.

Key Words

UO₂, Spark Plasma Sintering (SPS), thermal conductivity

1. Introduction

Uranium dioxide (UO₂) is the most common fuel material used in commercial nuclear power reactors. Compared to other fuel materials, UO₂ has the advantage of high melting point[1], good high-temperature stability, good chemical compatibility with both cladding and coolant and resistance to radiation[2]. However, the main drawback of a UO₂ fuel pellet is its extremely low thermal conductivity, which results in a fairly high centerline temperature during operation in a reactor and a steep radial temperature gradient, typically around a 500°K drop within a pellet radial distance of 5mm. Thermal stress induced by this large temperature gradient may lead to cracking within the fuel pellets and cause accelerated release of fission gases, both of which limit the operational life of UO₂ fuel in a reactor. Therefore, the importance of increasing the thermal conductivity of UO₂ fuel cannot be over emphasized.

The idea of incorporating high thermal conductivity material into a UO₂ matrix has been gaining popularity in recent years. However initial attempts to sinter UO₂ powder with second phase high conductivity materials such as SiC have been met with limited success, especially in terms of reaching the desired 96.5% of the theoretical density (TD) in the final pellet[3]. Ishimoto et al.[4] considered BeO as a second phase and sintered UO₂-BeO composite in a reducing atmosphere successfully. Most recently, numerous other second phase materials have been proposed in the literature. Yeo et al.[5, 6] and Cartas et al.[7]

utilized spark plasma sintering and fabricated UO_2 -SiC and UO_2 -carbon nanotube (CNT) composite fuel pellets, respectively. Lee et al.[8] proposed the idea of UO_2 -graphene composite and studied the thermal properties through numerical simulation. Among all the potential second phase materials, diamond has extremely high thermal conductivity (about 22 W/cm·K at room temperature), which is the highest of any solid material[9]. Due to its sp^3 hybridization bond structure, diamond is found to be resistant to the radiation environment[10]. Therefore, diamond makes an ideal second phase for incorporation into UO_2 pellets to achieve a higher thermal conductivity in the composite. From the economics perspective, the price of synthetically produced diamond powder is much lower (~\$2.5/g) than that of CNT (~\$270/g) or graphene (~\$125/g), which is beneficial for commercialization.

Fabrication of UO_2 fuel using conventional oxidative sintering methods requires elevated temperatures up to 1700°C for 2-4 hours. The entire temperature cycle (heating time, hold time, and cooling time) may take 10-15 hours due to slow heating rate of only around 5°C/min[11-13]. Ishimoto et al.[4] sintered UO_2 -BeO composite at 2073K for 2 hours using a conventional sintering method. However, using spark plasma sintering (SPS) technique, Yeo et al.[5] achieved high density UO_2 -SiC pellets at 1350°C with a hold time as low as 5 minutes. SPS is a promising approach for processing difficult-to-sinter powders such as UO_2 . Advantages of using SPS over traditional sintering of UO_2 can be found in the work of Ge et al[14]. The objective of this work is to fabricate UO_2 -

diamond fuel pellets using SPS and investigate the microstructure and properties of this novel composite fuel.

2. Experimental

2.1 Powder preparation

Figures 1 and 2 show SEM images of the as-received UO_2 and diamond powders, respectively. The uranium dioxide powder was obtained from AREVA Federal Services, Hanford, Washington, USA. The powder was reported to have a bulk density of 2.3g/cm^3 , mean particle diameter of $2.4\mu\text{m}$ and a BET surface area of $3.11\text{m}^2/\text{g}$. The grain size was determined to be 200-400nm, see Fig.1. The O/U ratio was determined to be 2.11 using ASTM equilibration method (C1430-07). The diamond powder was obtained from Advanced Abrasives, Pennsauken, NJ. Four sizes of diamond particles with a mean particle size of $0.25\mu\text{m}$, $3\mu\text{m}$, $12\mu\text{m}$ and $25\mu\text{m}$, were used in this study.

A SPEX-8000 shaker was used to blend UO_2 and a 5vol% diamond powder for 1 hour with a blending aid 2,3-Dihydroperfluoropentane. The use of a blending aid was proven to be effective and non-contaminative to the final powder[15].

Fig. 1. SEM image of as-received UO_2 powder

Fig. 2. SEM images showing morphologies of diamond powder with a mean particle size of a) $0.25\ \mu\text{m}$, b) $3\ \mu\text{m}$, c) $12\ \mu\text{m}$, and d) $25\ \mu\text{m}$.

2.2 SPS

A graphite die with inner diameter of 12.7 mm and outer diameter of 31.5 mm was used to sinter the mixed powder. A thin graphite foil was inserted into a graphite die and then 4g of UO_2 +diamond mixture was poured into the die. Graphite punches were used on either side of the die to hold the powder. The graphite foil prevents reaction between the powders and the die. A Dr. Sinter[®] SPS-1030 spark plasma system (SPS) machine was used to sinter the blended powder. The entire die assembly with powder and punches was placed in the SPS chamber. During sintering, a vacuum of less than 10 Pa was maintained in the chamber and the temperature was measured using a pyrometer that focused on the surface of the graphite die. A temperature ramp rate of 100°C/min was employed and the maximum sintering temperature was varied from 1300°C to 1600°C with a hold time of 5 minutes for various powder mixtures. An axial pressure of 36 MPa was applied when the maximum sintering temperature was reached, and the pressure was released when the cooling process started. More detailed description of this method can be found in recent publications[5, 6, 15, 16]. It is important to note that although the stoichiometry of the starting uranium dioxide powder was 2.11, the O/U ratio has changed to be 2.00 due to the reducing environment in SPS. Similar result was first observed in fabricating pure UO_2 pellets using SPS[14] where a detailed analysis regarding the change in stoichiometry in UO_2 during SPS can be found. Thus, SPS eliminates additional

post-sintering steps commonly used in conventional sintering method of UO₂ pellets.

2.3 Characterization methods

Upon cooling to room temperature, the pellets were removed from the graphite dies. The density of each sintered pellet was measured by the Archimedes method and converted to relative density. The theoretical density of the composite can be calculated from the rule of mixture, i.e.,

$$\rho_c = \rho_d V_d + \rho_{uo_2}(1 - V_d) \quad (1)$$

where ρ_c is the theoretical density of the composite, ρ_d , V_d and ρ_{uo_2} are density of diamond, volume fraction of diamond, and density of UO₂, respectively. The density and relevant values of diamond and UO₂ are listed in Table 1.

X-ray diffraction (XRD) measurements were conducted to investigate the chemical reactions which may have occurred between UO₂ and diamond during the sintering process. As the detection limit of XRD is around 5wt% of chemical compounds and the short processing time of 5 minutes may not be enough to form reaction compounds at sufficient quantity, the volume fraction of diamond was increased to 70% in the composite for this specific study only. The mixing process and sintering procedures were the same as the other pellets. X-Ray diffraction patterns were recorded with a PANalytical X'Pert Powder[®] diffractometer at room temperature. The 2θ angle was scanned from 20° to 80° with a step size of 0.005°. The software X'Pert HighScore Plus[®] was used for peak fitting.

To characterize the microstructure of the sintered pellets, scanning electron microscopy (JEOL-6335L and FEI XL-40 FEG-SEM) was used. The secondary electron mode was chosen for the images and the accelerating voltage was varied between 10 to 15 KV.

The thermal conductivity of the sintered composite pellets were calculated using equation (2)

$$K_c = \alpha c_p \rho_c \quad (2)$$

where α is the thermal diffusivity, c_p is the specific heat and ρ_c is the density of the composite. α was measured at three different temperatures, i.e., 100°C, 500°C and 900°C, using a laser flash method (Anter Flashline[®]-3000). Specific heat was calculated using the Neumann-Kopp rule[17]:

$$c_p = c_d f_d + c_{uo_2} (1 - f_d) \quad (3)$$

where c_d and c_{uo_2} are the specific heat of diamond and UO₂, respectively, and f_d is the weight fraction of diamond.

The Young's modulus was determined using the ultrasonic measurement method, an accurate and non-destructive technique. Using a pulser/receiver system (Model 5072PR, Olympus, Waltham, MA), both longitudinal velocity V_L and shear velocity V_s of the pellets were measured. The Young's modulus is then calculated by the following equation[18]:

$$\nu = \frac{1-2(\frac{V_S}{V_L})^2}{2-2(\frac{V_S}{V_L})^2} \quad (4)$$

$$E = \frac{V_L^2 \rho (1+\nu)(1-2\nu)}{1-\nu} \quad (5)$$

where ν is Poisson's ratio, E is Young's modulus and ρ is the density of the pellet.

Four measurements were performed on each composite and the average value was calculated.

Table 1. Properties of UO₂ and diamond.

Materials	Density (g/cm ³)	Specific heat, c_p (J/Kg•K) [19, 20]		
		100°C	500°C	900°C
UO ₂	10.96	258	305	314
Diamond	3.52	757	1687	1849

3. Results and discussion

3.1 Density

Density has a huge influence on the thermal and mechanical properties of the composites. Therefore it is important to obtain high-density composite pellets.

Figure 3 shows the variation in the relative density of the UO_2 -diamond composites as a function of sintering temperature and diamond particle size. A horizontal line at 96.5% of the theoretical density (TD), which is the requirement for a fuel pellet to be used in a nuclear reactor[21], is plotted as the reference density. For all diamond particle sizes, the density of the sintered pellets increased with increasing sintering temperature up to 1500°C . While when the maximum sintering temperature achieve 1600°C , a slightly decrease in density can be observed. The decrease may be due to the formation of micro-cracks and the chemical reaction at high temperature, which will be discussed in the following sections. For nano-size diamond particles ($0.25\ \mu\text{m}$), the pellet density is consistently lower than other composites sintered under the same sintering conditions. A pellet with a 96.5% of the TD cannot be achieved even at a temperature of 1600°C . While for micro-size diamond particles ($3\ \mu\text{m}$, $12\ \mu\text{m}$, and $25\ \mu\text{m}$), high density pellets can be achieved when the maximum sintering temperature is 1400°C or above. The poor densification behavior of UO_2 - $0.25\ \mu\text{m}$ diamond composite could be explained by the microstructure of the sintered pellet, which will be discussed later.

Fig.3. The influence of sintering temperature and diamond particle size on the density of $\text{UO}_2+5\text{vol}\%$ diamond composite fuel

3.2 Chemical Reaction

Understanding the potential chemical reactions between diamond and UO_2 during SPS is critical to an evaluation of the performance potential of the composite pellet in the reactor environment. The chemical reaction products, (if any) may badly influence the thermal and mechanical performance of the sintered pellets. Figure 4 shows the XRD spectra of UO_2 and diamond powder as well as the UO_2 -Diamond composite at four different temperatures. For the spectra of starting powders, all characteristic peaks of UO_2 (Powder Diffraction

File (PDF) 00-005-0550) and diamond (PDF 01-075-0223) were clearly detected, revealing the cubic structure of UO_2 and diamond. The crystal planes were identified and marked in the plot. For the UO_2 -70 vol% diamond composite sintered at 1300°C , no other peaks, except UO_2 and diamond peaks, were observed. However, when the maximum sintering temperature was increased to 1400°C , a clear graphite peak was detected. The formation of graphite may be an indication of the graphitization of diamond. Although it is known that the graphitization rate of diamond is extremely slow below 1600°C in an inert gas environment[22], this temperature could decrease significantly if high pressure is applied. During SPS, a pressure of 36 MPa was applied at the maximum sintering temperature, which could facilitate the graphitization process. When the maximum sintering temperature reached 1500°C or higher, not only graphite, but also uranium carbide (UC) peaks were observed. The following reactions between UO_2 and carbon are expected[23]:

Fig. 4. XRD spectra of starting powder and UO_2 -70 vol% diamond pellets sintered at various temperatures revealing formation of UC and graphite phases at higher temperature.

Note that there is CO gas formed in these reactions. Recall that slightly decrease in density was observed in pellets sintered at 1600°C , which may be due to the formation of gas. During the hold time at such high temperature, the sintering process was almost done, but some micro holes were left because of the formation of CO. The increase of porosity thus led to a decrease in final density. Clearly, higher sintering temperatures are not preferable due to the formation of graphite and other reaction products. Sintering temperatures well below 1500°C are recommended for fabrication of UO_2 -diamond composite pellets.

3.3 Microstructure

3.3.1 Dispersion of diamond particles

The dispersion of diamond particles in the composite pellets sintered at 1400°C can be seen in Fig. 5. The diamond particles with mean size of 3 μm , 12 μm and 25 μm were seen to be relatively uniformly distributed in the UO_2 matrix from Figs. 5(b)-5(d), while the agglomeration of 0.25 μm nano-size diamond particles was observed in Fig. 5(a). The agglomeration could be due to the poor mixing. Longer mixing time (6 hour) was also attempted but no obvious improvement was observed. Therefore, the current milling process is proven not to be an appropriate approach for mixing nano sized diamond in UO_2 powder. The agglomeration of diamond particles may be a reason for the poor densification observed in Fig. 3 in UO_2 -diamond composite with a diamond particle size of 0.25 μm . In the region of diamond agglomeration, UO_2 -diamond interfaces were replaced instead by diamond-diamond interfaces. Because the diamond is the most covalently bonded material, the sintering of diamond particles is much more difficult. As a result, the pellets did not sinter fully and loose diamond powder regions were seen in the microstructure of UO_2 -0.25 μm composite pellet, see Fig 5(a). This led to a poor measured density in the sintered nano-diamond composite pellets as well.

Fig. 5. Fracture surfaces of UO_2 -diamond pellets sintered at 1400°C with diamond particles of size (a) $0.25\ \mu\text{m}$, (b) $3\ \mu\text{m}$, (c) $12\ \mu\text{m}$, and (d) $25\ \mu\text{m}$. Note that agglomeration of diamond particles is seen in (a).

3.3.2 UO_2 -diamond interface

The UO_2 -diamond interfaces in various UO_2 -diamond composites are shown in Fig. 6. The diamond particles had good interfacial contact with the UO_2 matrix in pellets containing $0.25\ \mu\text{m}$, $3\ \mu\text{m}$ and $12\ \mu\text{m}$ diamond particles, but several micro-cracks were observed in pellets containing $25\ \mu\text{m}$ diamond particles. The formation of micro-cracks were due to a mismatch in coefficients of thermal expansion (CTE) between the UO_2 and the diamond particles. It is noted that the CTE of UO_2 and diamond is $9.93 \times 10^{-6}\ \text{K}^{-1}$ and $1 \times 10^{-6}\ \text{K}^{-1}$ at room

temperature, respectively. Although the CTE would slightly change with temperature, a large difference in CTE between UO_2 and diamond existed at all temperatures[24, 25]. Due to this mismatch, diamond particles were subjected to a large compressive stress during the cooling process. The larger the particle size was, the greater the compressive stress would be. Micro-cracks are initiated from the interface when the **tensile stress in UO_2** exceeds a critical value in the UO_2 matrix. Similar phenomena were also seen in UO_2 -SiC composites where such CTEs mismatch existed between matrix and second phase particles[6]. The induced micro-cracks adversely influence the thermal and mechanical properties of the composite pellets, which is discussed next.

Fig. 6. UO_2 -diamond Interface in composite pellets sintered at 1400°C with mean diamond particle size of a) 0.25 μm , b) 3 μm , c) 12 μm , and d) 25 μm .

3.3.3 Grain size

Figure 7 reveals the polished and thermally-etched surfaces of UO_2 -diamond composite pellets sintered at 1400°C with a hold time of 5 minutes. Uniform grain sizes were revealed in pellets with $3\ \mu\text{m}$ diamond ($2.6\ \mu\text{m}$, Fig. 7(d)), $12\ \mu\text{m}$ diamond ($2.9\ \mu\text{m}$, Fig. 7(3)) and $25\ \mu\text{m}$ diamond ($3.1\ \mu\text{m}$, Fig. 7(f)), while an obvious non-uniform grain size distribution was observed in the UO_2 - $0.25\ \mu\text{m}$ diamond pellet (Fig. 7(a)-7(c)). Typical UO_2 grain sizes were no larger than $3.5\ \mu\text{m}$ in all these microstructures. However, in the work of Ge et al[14], it was shown that the grain size of UO_2 pellets sintered by SPS can be as large as $8.9\ \mu\text{m}$, much larger than the current UO_2 grain sizes in UO_2 -diamond composites. The results are reasonable when the pinning effect of second phase particles are considered. During sintering, the UO_2 grains start to grow and once grain grain boundary reaches a diamond particle, the grain boundary migration was pinned by that particle. Therefore, smaller grains were formed in the UO_2 matrix. Since the volume fraction of diamond is the same in all composites, a higher surface to volume ratio of diamond particles exists in composite containing smaller diamond particles. Therefore, the intensity of pinning effect would be more in composite with $0.25\ \mu\text{m}$ diamond. It can be seen that within these four UO_2 -diamond composites, the pellet with $25\ \mu\text{m}$ diamond particles has the largest grain size, which is in agreement with the above argument.

The formation of the non-uniform grain size in Fig. 7(a) can be owing to the uneven distribution of nano diamond particles. Recall that in Fig 5(a), the

agglomeration of diamond particles was observed. Therefore, the region A which lacks diamond particles has much larger grain size ($2.5\ \mu\text{m}$) than the region B ($0.7\ \mu\text{m}$) which has well distributed diamond particles.

Fig. 7 Polished and thermally etched surfaces of UO₂-diamond composites with mean diamond particle size (a) $0.25\ \mu\text{m}$, (d) $3\ \mu\text{m}$, (e) $12\ \mu\text{m}$, and (f) $25\ \mu\text{m}$. (b) and (c) are

enlarged pictures of areas in a). The grain sizes are indicated on the top left of each image.

3.4 Thermal conductivity

As discussed in the previous section (Eq. (2)), the thermal conductivity is a product of thermal diffusivity, specific heat and pellet density. The specific heat of UO_2 -5 vol% diamond composite was calculated, using Eq. (3) and the values listed in Table 1, to be 266.3, 327.5 and 339.7 J/kg·K at 100°C, 500°C and 900°C, respectively, and then the thermal conductivity was calculated. Figure 8 shows the thermal conductivity for UO_2 -diamond composites sintered at 1400°C with various sizes of diamond particles. The dashed lines represent the thermal conductivity of pure UO_2 in the literature at the specified temperatures.

Compared to pure UO_2 , an increase in thermal conductivity was observed for composite pellets which containing 0.25 μm , 3 μm or 12 μm diamond particles, on the contrary, the pellets which contained 25 μm diamond showed a lower thermal conductivity than pure UO_2 . The maximum increase in thermal conductivity was observed in pellet containing 3 μm diamond particles, which is found to be 41.6%, 38.3% and 34.2% at 100°C, 500°C and 900°C, respectively.

For particle reinforced composites, several theoretical models were utilized for estimating the effective thermal conductivity[26, 27]. Hasselman and Johnson's model[27], which has been proven to be effective by many researchers, is given by the following equation:

$$\lambda_c = \lambda_m \frac{[2(\frac{\lambda_d}{\lambda_m} - 1 - \frac{\lambda_d}{a \cdot h})V_d + (\frac{\lambda_d}{\lambda_m} + \frac{2\lambda_d}{a \cdot h} + 2)]}{[(1 - \frac{\lambda_d}{\lambda_m} + \frac{\lambda_d}{a \cdot h})V_d + (\frac{\lambda_d}{\lambda_m} + \frac{2\lambda_d}{a \cdot h} + 2)]} \quad (7)$$

where λ is the thermal conductivity, subscripts c , d and m are composite, particle and matrix, respectively, V_d is the volume fraction of particles, a is the radius of particle, and h is the interfacial thermal conductance. If the volume fraction of particles is fixed, it is easy to prove that the thermal conductivity of the composite is a monotonically increasing function of $a \cdot h$. Theoretically, the interfacial thermal conductance between matrix and second phase particles is related to the density and phonon velocity of the two materials[28]. Therefore, the interfacial thermal conductance between UO_2 and diamond is assumed to be a constant. The thermal conductivity, then, would increase with the increase in diamond particle size based on this model. However, the experimental values in Fig. 8 show a different trend. Pellets with 12 μm diamond and 25 μm diamond showed lower thermal conductivity than the UO_2 -3 μm diamond pellet. This result is because of the imperfections in the microstructure. Recall from Fig. 6(c) and 6(d) that at the interfaces between the UO_2 and diamond of sintered pellets, micro-cracks were observed in UO_2 -12 μm diamond and UO_2 -25 μm diamond composite. These micro-cracks have an adverse influence on the thermal conductivity. When the diamond particle size is larger than 12 μm , more cracks were formed and result in even lower thermal conductivity values.

A comparison of the thermal conductivity of the UO_2 -diamond composite and other UO_2 -based composites published in literature is shown in Fig. 9. The density of these pellets were not normalized to the same value because a density

of 96%-98% was achieved in all the pellets discussed. Among these composites, UO_2 -diamond was found to have the highest thermal conductivity at the same measured temperature.

Fig. 8 Thermal conductivity UO_2 -diamond pellets with various sizes of diamond particles. The dotted lines refer to UO_2 literature values[19] at each temperature.

Fig. 9 Comparison of thermal conductivity of UO₂ and various UO₂-based composites

3.5 Young's modulus

Although an increase in the thermal conductivity is the primary motive for fabricating UO₂-diamond composite pellets, the mechanical property of the pellets is also important for a successful application in nuclear reactors. Young's modulus of UO₂-diamond composites, therefore, is measured and compared with pure UO₂ pellets.

Figure 10 shows the Young's modulus of UO₂-5 vol% diamond composite pellets with different diamond particle sizes. All pellets were sintered at 1400°C with a hold time of 5 minutes. The values of pure UO₂ pellets from different

publications are also plotted for comparison. Note that Gatt et al[29], and Padel and Noyion[30] fabricated the pellets using conventional sintering method, while Ge et al[16] used spark plasma sintering (SPS) technique to sinter UO_2 pellets. Due to the good agreement between their results, it has been concluded that the sintering technique has no influence on the Young's modulus of UO_2 pellets. The UO_2 -diamond composites are expected to have higher Young's modulus than pure UO_2 due to the extremely high Young's modulus of diamond (1220 GPa) and pellets with different particle sizes should theoretically have the same value if the diamond concentration is the same. However, it is seen in the figure that only pellets with the 3 μm diamond particles revealed a higher Young's modulus than the pure UO_2 pellets, while the pellets with 0.25 μm , 12 μm and 25 μm diamond showed a much lower value. It is known that the Young's modulus is highly sensitive to the micro-cracks, therefore, the results are consistent with the observation of micro-crack presence in those pellets. For UO_2 -0.25 μm diamond pellet, no cracks were revealed by SEM (Fig. 6(a)). The low Young's modulus for this composite may be due to its low density compared to other pellets. The variation in Young's modulus is consistent with the thermal conductivity results shown in Fig.8. Clearly, both thermal and mechanical properties are strongly influenced by the formation of micro-cracks, and therefore the significance of avoiding them is self-evident.

Fig.10. Young's modulus of UO₂-diamond composites as a function of mean particle size of diamond. Note that arrays represent the Young's modulus of pure UO₂ pellets reported from literature.

From the above experimental and characterization results, it is seen that 3 μm diamond particles provide optimal density, microstructure, thermal conductivity, and mechanical property. Such composites are candidate materials for accident tolerant fuel because those pellets can operate at lower temperatures, reduced fission gas release and can dissipate residual heat more quickly in a loss-of-coolant accident (LOCA) in nuclear reactor. For the design of UO₂-diamond composite pellet, the content of diamond is to be optimized in the future. It is a trade-off because more diamond may theoretically give a better thermal conductivity but lower nuclear reactivity due to the reduction in UO₂

content in the pellet. Therefore, more studies and calculations are still needed. In addition, our future research will include investigation of mechanical integrity and chemical stability of these composite pellets at long-duration high temperature exposure and eventually testing their effectiveness in advanced test reactor at Idaho National Lab.

4. Conclusion

The concept of high thermal conductivity UO_2 -diamond composite fuel pellets has been demonstrated. Using the spark plasma sintering technique, UO_2 -5 vol% diamond composite fuel pellets were successfully sintered. A maximum sintering temperature of 1400°C and a hold time of 5 minutes are recommended in order to achieve a high density pellet with minimal UO_2 -diamond chemical reaction. Four diamond particle sizes ranging from $0.25\ \mu\text{m}$ to $25\ \mu\text{m}$ were investigated. However, pellets with $0.25\ \mu\text{m}$ diamond particles were not acceptable due to the poor mixing and the resulting low densification. On the other hand, micro-cracks were observed when the diamond particle size was larger than $12\ \mu\text{m}$, which adversely influenced both the thermal and mechanical properties of the composites. Therefore, a $3\ \mu\text{m}$ diamond is recommended for the UO_2 -diamond composite pellet in order to obtain the best thermal and mechanical performance.

For the UO_2 -diamond composite pellets, the maximum increase in thermal conductivity was found to be 41.6%, 38.3% and 34.2% at 100°C , 500°C and

900°C, respectively. These results are higher than those reported in literature for other UO_2 -based composites. Therefore, diamond could probably be a more effective candidate for UO_2 composite fuel pellets.

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Fig1

Fig2a

Fig2b

Fig2c

Fig2d

Fig3

Fig4

Fig5a

Fig5b

Fig5c

Fig5d

Fig6a

Fig6b

Fig6c

Fig6d

Fig7a

0.25 μm diamond
Bimodal distribution

A: $2.5 \pm 0.4 \mu\text{m}$

B: $0.7 \pm 0.03 \mu\text{m}$

A

B

Acc.V	Spot	Magn	Det	WD	Exp
10.0 kV	5.0	5000x	SE	13.6	0

10 μm

Fig7b

Fig7c

Fig7d

Fig7e

Fig7f

Fig8

Fig9

Fig10